Support Information

Experiment Section:

1. Materials

Triethoxy-3-(2-imidazolin-1-yl) propylsilane (IZPES) was purchased from Sigma Aldrich. Phenyl-triethoxysilane (PTES) was purchased from Acros. Tetraethyl orthosilicate (TEOS), 1-chlorobutane, Potassium hexafluorophosphate (KPF_6), sodium fluoroborate ($NaBF_4$) and bistrifluoromethanesulfonimide lithium [$Li(CF_3SO_2)_2N$] were purchased from Sinopharm Chemical Reagent Beijing Co. (SCRC). Hydrolyzed styrene-maleic anhydride (HSMA) copolymer was synthesized.[S1]

2. Characterization:

Morphology of the samples was characterized via scanning electron microscopy (SEM) HITACHI S-4800 operated at 15 kV. The samples for SEM observation were prepared by vacuum sputtering with Pt after being ambient dried. FT-IR spectroscopy was performed after scanning the samples for 32 times using a Bruker EQUINOX 55 spectrometer with the sample/KBr pressed pellets. Elemental measurement was performed on X-ray photoelectron spectroscopy ESCALab220i-XL (VG Scientific) using 300W AlKα radiation. Morphology of the emulsions stabilized with the IL-Janus nanosheets was observed under Leica DMLP microscope. Zeta potential is measured with Malvern Nanosizer ZS-90.

3. Synthesis of Imidazolin Based Janus Nanosheets:

15 mL of 10% hydrolyzed styrene-maleic anhydride (HSMA) solution was dissolved in 75 mL of water. The solution was kept at 70 °C after pH of the mixture was adjusted to 3.0 with 2M hydrochloric acid. At 70 °C, 25.0 g of paraffin (Tm: 52-54 °C) was mixed with 5.2 g of TEOS (tetraethylorthosilicate), 1.37 g of IZPES (triethoxy-3-(2-imidazolin-1-yl)propylsilane) and 1.2 g of PTES (phenyltriethoxysilane) under stirring. The oil mixture was dispersed into the aqueous solution with a homogenizer at a speed of 13,000 rpm for 5 min, forming an oil-in-water emulsion. The emulsion stood at 70 °C for 12 h for a self-organized sol-gel process at the emulsion interface. After the resultant emulsion was cooled down to room temperature, the core/shell particles with a paraffin core were obtained by a sequential filtration and wash with water. Afterwards, the particles were immersed in THF to dissolve the paraffin core under ultrasonication, forming imidazolin based Janus hollow particles. Imidazolin based Janus nanosheets were obtained by crushing the Janus hollow particles with colloid milling.

4. Synthesis of Ionic Liquid (IL) Based Janus Nanosheets:

After 100 mg of Imidazolin based Janus nanosheet was dispersed in 40 mL of toluene, 1 mL of 1-chlorobutane was added and refluxed for 24 h. The modified nanosheets were separated after centrifugation and wash with toluene for several times to remove residual 1-chlorobutane, and vacuum dried. Cl^- based ionic liquid (IL) Janus nanosheets were obtained as a yellow powder.

5. Anion Exchange Process of IL-Janus Nanosheets:

After 30 mg of Cl^- based IL-Janus nanosheet was dispersed in 10 mL of water, 10 mg of KPF_6 was added and dissolved under ultrasonication for 1 min. The system stood under stirring at room temperature for 1 h. The PF_6^- based IL-Janus nanosheet was obtained after centrifugation and

wash with water for several times. Similarly, the corresponding IL-Janus nanosheets with varied anion groups were prepared when using other salts for example $NaBF_4$, $Li(CF_3SO_2)_2N$ and $NaHSO_4$.

6. Emulsification with the Janus Nanosheets:

10 mg of the Cl^- based IL-Janus nanosheets was added in a glass bottle containing 1 mL of water (a trace amount of chromogenic agent methyl orange was added into water) and 1.5 mL of decane. An oil-in-water emulsion was formed after the mixture was shaken.

At 70 °C, 80 mg of the Cl^- based IL-Janus nanosheets was added in a glass bottle containing 3 g of water and 1 g of paraffin (Tm: 52-54 °C). After the mixture was shaken, a melt paraffin-in-water emulsion was formed. The emulsion was naturally cooled down to room temperature to let the paraffin core crystallize.

14 mg of the Cl^- based IL-Janus nanosheets was added in a glass bottle containing 2 mL of water (a trace amount of chromogenic agent methyl orange was added into water) and 1.2 mL of toluene. A water-in-oil emulsion was formed after the mixture was shaken.

S1. Z. G. Jin, Y. Wang, J. G. Liu, Z. Z. Yang, *Polymer* **2008**, 49, 2903-2910.

Supporting Figures and Tables:

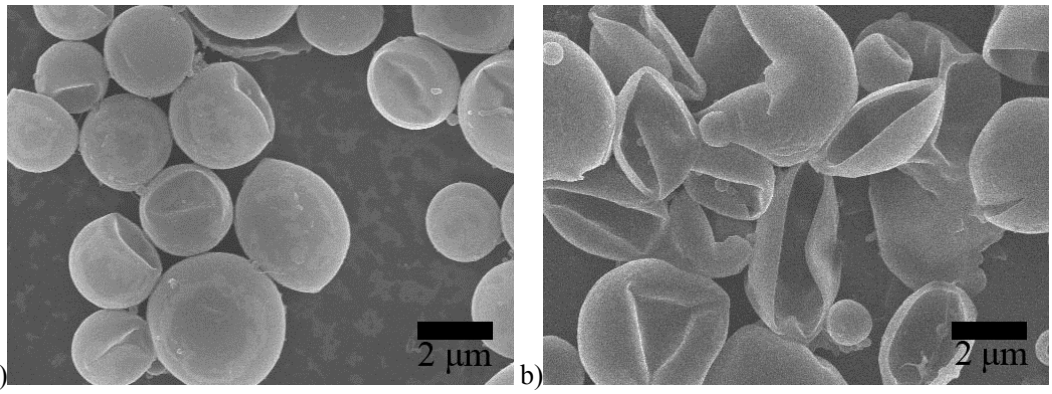


Fig. S1. a) SEM image of the core/shell particles after being cooled to room temperature; b) SEM image of the hollow particles after the paraffin core is dissolved.

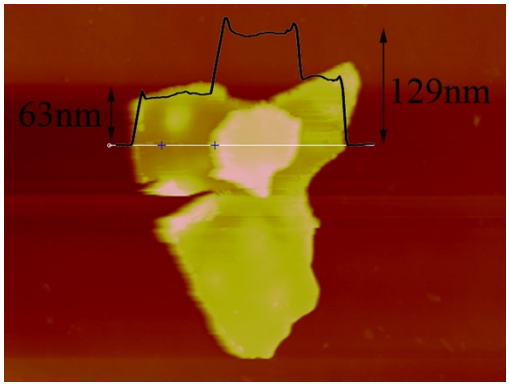


Fig. S2. AFM image of the representative silica Janus nanosheets.

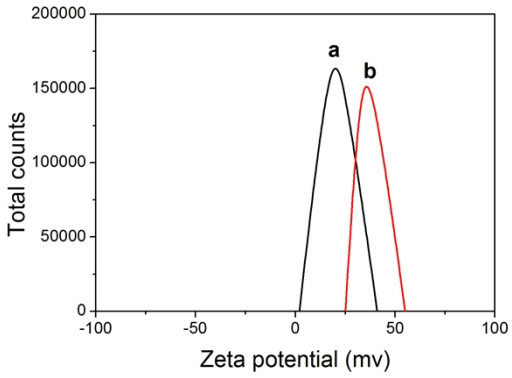


Fig. S3. Zeta potential of imidazolin (a) and Cl^- IL (b) based Janus nanosheets measured at pH=7.0.

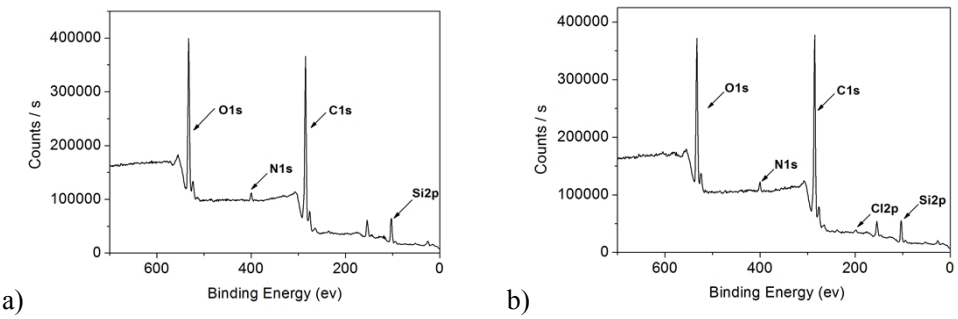


Fig. S4. XPS spectra of the imidazolin (a) and Cl^- IL (b) based Janus nanosheets.

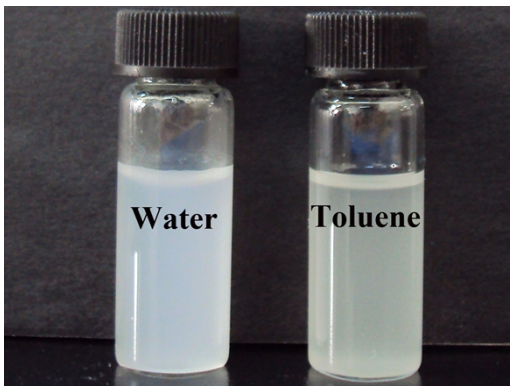
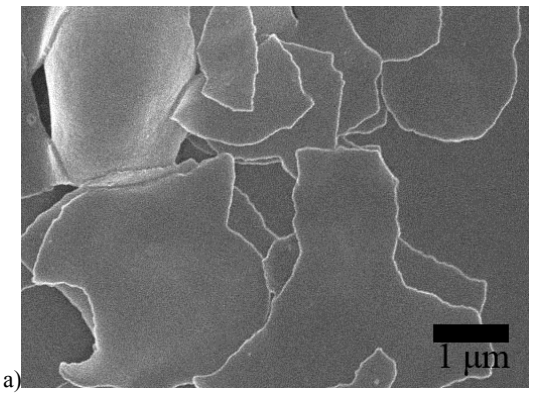
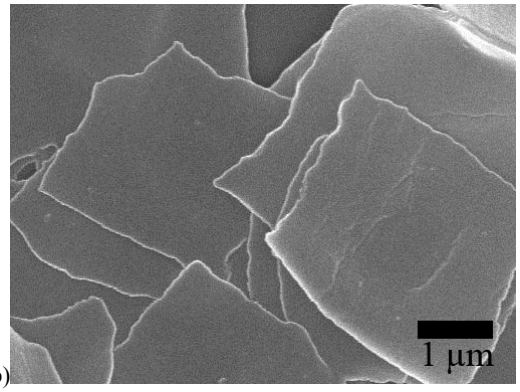
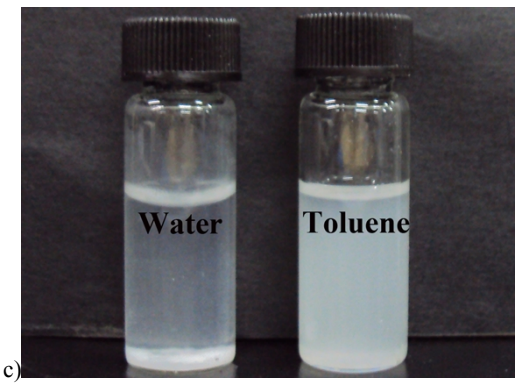
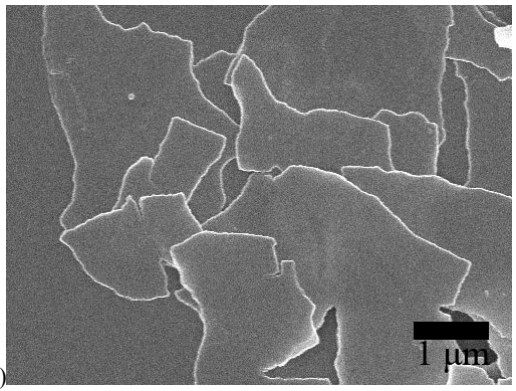


Fig. S5. Dispersions of the Cl^- based IL-Janus nanosheets in water (left) and toluene (right).









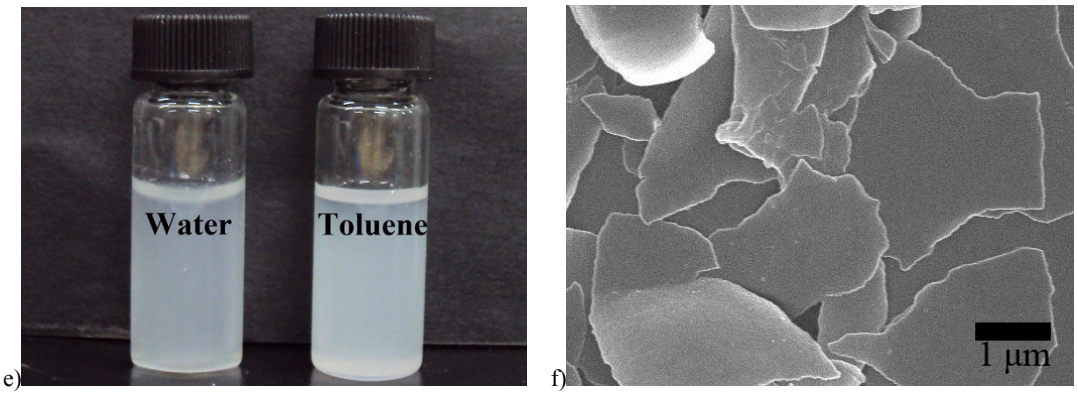


Fig. S6. a) SEM image of the PF_6^- based IL-Janus nanosheets after drying from ethanol dispersion; b) SEM image of the BF_4^- based IL-Janus nanosheets after drying from ethanol dispersion; c) $(CF_3SO_2)_2N^-$ based IL-Janus nanosheets aggregated in water (left) and dispersed in toluene (right); d) SEM image of the $(CF_3SO_2)_2N^-$ based IL-Janus nanosheets after drying from ethanol dispersion; e) dispersions of the HSO_4^- based IL-Janus nanosheets in water (left) and toluene (right); f) SEM image of the HSO_4^- based IL-Janus nanosheets after drying from ethanol dispersion.

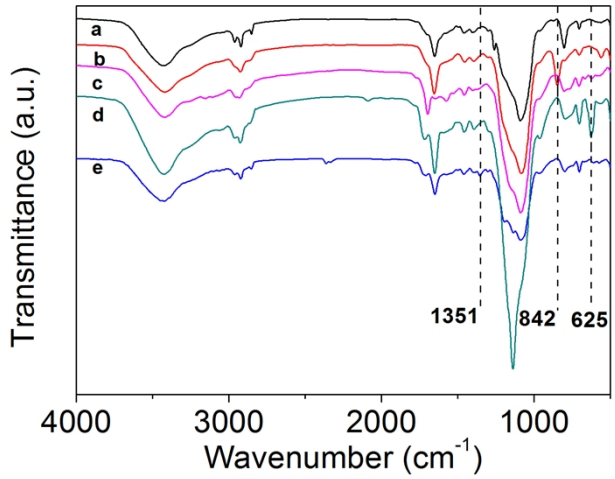


Fig. S7. FT-IR spectra of some representative IL-Janus nanosheets with varied anion groups: a) Cl^-; b) PF_6^-; c) BF_4^-; d) HSO_4^-; e) $(CF_3SO_2)_2N^-$.

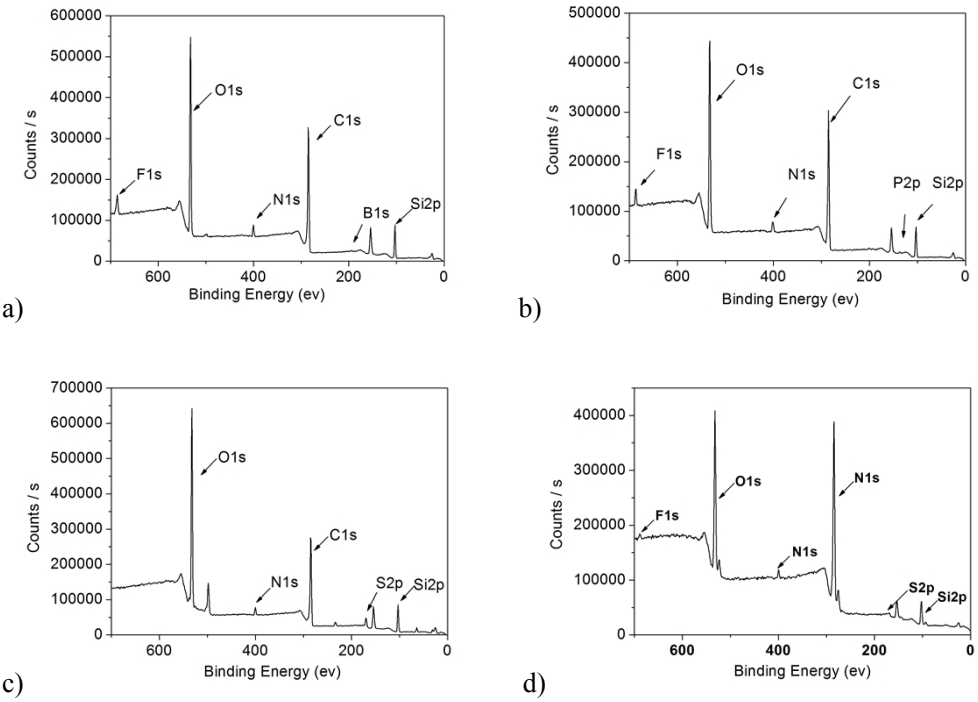


Fig. S8. XPS spectra of some representative IL-Janus nanosheets with varied anion groups: a) BF_4^-; b) PF_6^-; c) HSO_4^-; d) $(CF_3SO_2)_2N^-$.

Table S1. Elemental analysis of the Janus nanosheets by XPS measurement after feeding varied amount of KPF_6.

No.	*Cl^- based IL Janus nanosheets (mg)*	*KPF_6 (μmol)*	*Cl (atomic %)*	*P (atomic %)*
1	1.0	0.0	100	0.0
2	1.0	2.0	90.2	9.8
3	1.0	4.0	49.9	50.1
4	1.0	4.5	19.4	80.6
5	1.0	5.0	8.6	91.4
6	1.0	5.5	1.6	98.4
7	1.0	6.0	0.0	100

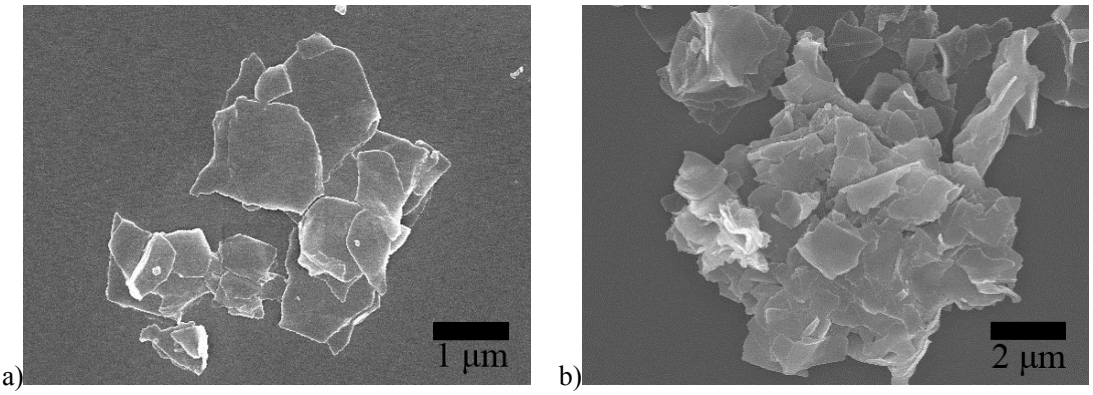


Fig. S9. SEM images of the Cl^- based IL-Janus nanosheets after drying from aqueous dispersion before a) and after b) feeding 4.5 μmol of KPF_6. All the systems stand for 10 min.

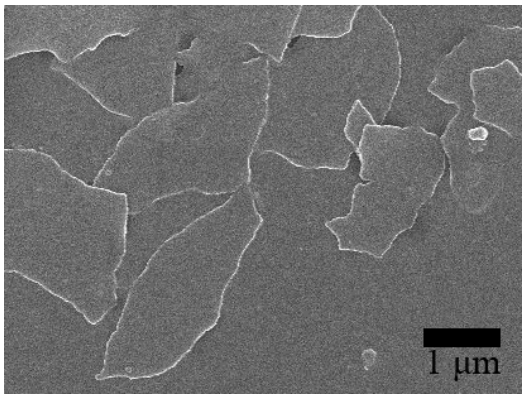


Fig. S10. SEM image of the PF_6^- IL based Janus nanosheets after drying from the dispersion in toluene.

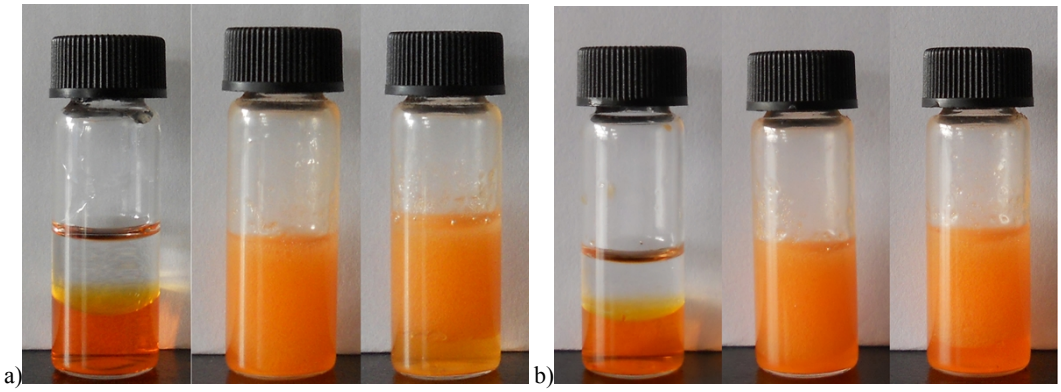


Fig. S11. a) Immiscible mixture of decane (top) and water (bottom) (left), the oil-in-water emulsion stabilized by the imidazolin based Janus nanosheets (decane/water 3:2 vol/vol) (middle), and the emulsion after feeding 10 μmol of KPF_6 (right); b) immiscible mixture of decane (top) and water (bottom) (left), the oil-in-water emulsion stabilized with the Cl^- based IL-Janus nanosheets (decane/water 3:2 vol/vol) (middle), and the emulsion after feeding 10 μmol of KCl (right).

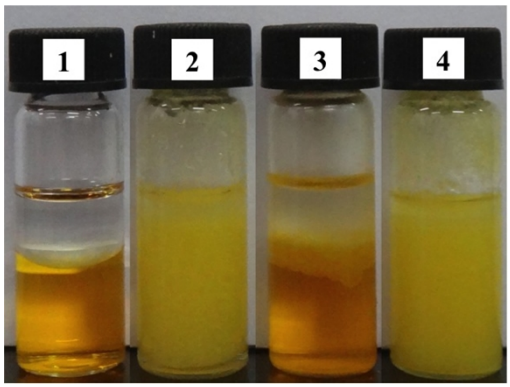


Fig. S12. Janus performance of the IL-nanosheets as anion responsive surfactants in a water-in-oil emulsion: 1) immiscible mixture of toluene (top) and water (bottom), methyl orange is added to water as chromogenic agent; 2) water-in-oil emulsion stabilized with the Cl^- based IL-Janus nanosheets after stirring, water/toluene volume ratio is 5:3; 3) feeding 10 μmol of KPF_6 in the emulsion (2) after stirring; 4) feeding Cl^- again in the emulsion (3) after stirring.